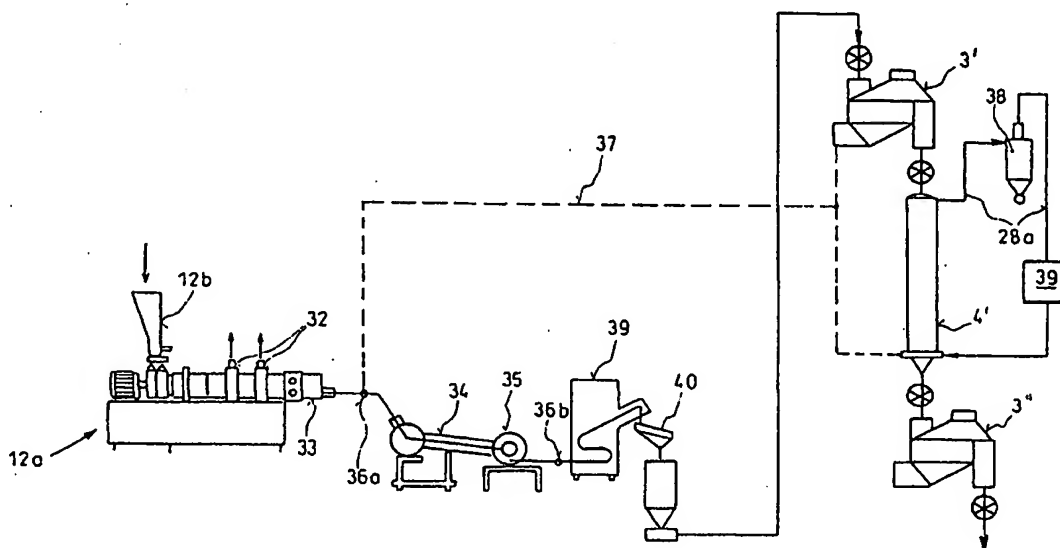




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(21) International Application Number: PCT/CH99/00580 (22) International Filing Date: 3 December 1999 (03.12.99) (30) Priority Data: 09/244,630 4 February 1999 (04.02.99) US (71) Applicant (for all designated States except US): BÜHLER AG [CH/CH]; Patentabteilung, CH-9240 Uzwil (CH). (72) Inventors; and (75) Inventors/Applicants (for US only): BORER, Camille [CH/CH]; Hellerweg 12, CH-8247 Flurlingen (CH). CULBERT, Brent-Allen [NZ/CH]; Marktgasse 66, CH-9500 Wil (CH).		(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i>

(54) Title: PROCESS FOR UPGRADING PLASTIC MATERIAL**(57) Abstract**

In a process for upgrading cleaned used plastic material to be reclaimed, all steps are performed in a continuous way so as to extrude continuously the plastic material to provide elongated strands thereof, cutting the strands continuously as they are supplied by extrusion to convert them into pellets. Then, the pellets are fed without any intermediate storage to a continuous crystallizer in a stream of hot gas to heat the pellets to crystallizing temperature.

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PROCESS FOR UPGRADING PLASTIC MATERIAL

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Field of the Invention

This invention relates to process of reclaiming and upgrading cleaned used plastic material, particularly polyester material, such as polyethylene terephthalate. More particularly, the invention relates to process where used plastic material, and particularly polyethylene terephthalate, is upgraded to bottle grade material. When reclaiming, cleaning may be done before or within the framework of the present process. More specifically, the present invention relates to a process for upgrading cleaned used plastic material to be reclaimed comprising the steps of continuously extruding used plastic material to provide elongated strands, continuously cutting the strands to convert them into pellets, and feeding said pellets to a crystallizer.

Background of the invention

U.S. Patent No. 5,225,130 or WO 98/40194 disclose such a process. Typically, this known process, particularly that of the WO document, provides melting and cutting the cleaned and sorted plastic material which steps can be done in an extruder followed by and cooperating with an extruder. Extruding is a typical continuous step. After this continuous step, however, both documents suggest crystallizing and condensing the material in a tumbler or similar device under vacuum in solid state.

This known process has a series of disadvantages. It is known that tumblers are often provided with a vacuuming facility so that such tumblers are the obvious device for carrying out the process. However, good sealing is a precondition, because any vacuum draws air from the environment in, especially if the seal used becomes worn out or damaged. In such a case, oxygen from air can affect the quality of the reclaimed end product.

Moreover, tumblers can only be operated in batches. This leads to the necessity of an intermediate storage facility which involves additional investment costs and the requirement of additional space. Furthermore, the production capacity of tumblers and like

devices operating in batches is very limited so as to inhibit reclaiming large quantities of plastic material. Since, however, plastic material, such as PET, is increasingly in use, there is a necessity of reclaiming large quantities which cannot be done with the existing batch equipment. All these drawbacks, i.e. intermediate storage and relative small quantities handled at a necessarily high investment, result, in addition, in relative high reclaiming costs.

A further problem with PET or other plastic material to be recycled is that it may be contaminated by some chemicals which migrated in it during the former use. A typical example of such chemicals are aromatic compounds or flavours which stem from beverages or other liquids in those bottles. Another example are acids, soaps, and the like which contaminated the PET material after ordinary use. Adherents from labels have also to be removed. The prior art paid little or no attention to these factors which are relevant when it is intended that the reclaimed material should have bottle grade quality.

In this connection, reference should also be made to German patent publication No. 198 54 689, the contents of which being incorporated herein.

Summary of the invention

Therefore, it is an object of the present invention to devise an improved process of the kind described which leads to a higher yield being capable of reclaiming large quantities of used plastic material.

It is another object of the invention to reduce investment costs and the expenses of operation in a process of the kind described, particularly by avoiding the necessity of intermediate storage.

It is a further object of the present invention to find a process enabling to produce a better quality of reclaimed plastic material.

These objects are substantially achieved by using continuous treatment steps throughout the whole process and, more particularly, continuously crystallizing the pellets, as they are fed after cutting, in a stream of hot gas to heat the pellets to crystallizing temperature.

According to a preferred embodiment a continuous after-condensation or heating step is carried out after crystallizing.

- 5 When using a continuous fluidized bed, which is preferred according to the invention, it is most favourable if this fluidized bed has a plunger characteristic which means that all the pellets are treated during a substantially uniform period of dwell within the fluidized bed. A previous whirling bed, as in the prior art, can be omitted, although the present invention does not prevent using it depending on the stickiness of the product.

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It is most favourable if fluidisation or other treatment is done in an atmosphere of an inert gas, such as nitrogen, so as to avoid penetration of air or oxygen into the equipment, because the pressure of the inert gas will prevent it. In this way, not only the object of reducing costs and treating large quantities of plastic material is attained, but also
15 a higher quality of the end product.

Brief description of the drawings

Further details, characteristics and advantages will become apparent from the following description of a preferred example according to the invention schematically illustrated in
20 the drawings in which

Fig. 1 shows the cycle of PET bottle material;

Fig. 2 illustrates the process according to the invention as a detail of the cycle of Fig. 1,
and

- 25 Fig. 3 represents a practical example of an installation for recycling and decontaminating recycled PET material.

Detailed description of the drawings

According to Fig. 1, PET is produced in melting and pellet forming step 1 (normally an
30 extrusion step) after which the material is precrystallized in a step 2, preferably in a manner as described in U.S. Patent Nos. 5,091,134 or 5,119,570, i.e. in a whirling fluidized bed where the pellet's surface is dried as much as to avoid agglomerating and sticking of individual pellets in the following step. Step 2 is followed by a continuous af-

ter-crystallizing or main crystallizing step 3 in which, according to U.S. Patent Nos. 5,091,134 or 5,119,570, the pellets are fed with a plunger characteristic, i.e. a characteristic by which each particle has the same period of dwell in the fluidized bed and, therefore, is uniformly treated.

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After crystallizing in step 3, the PET material is polycondensed in a step 4 in a manner known *per se* and bottles are formed, normally by blow molding. The, thus, formed bottles are sold in step 5 to individual consumers after which they are either discarded (arrow 6) or are reclaimed (arrow 7).

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For reclaiming a collection system 8 is needed after which the collected material has to be washed, sorted and cleaned (step 9). Then begins the real reclaiming process 10 which can comprise a drying step 11 (sometimes necessary after the cleaning step 9 depending on the moisture of the material), a melting and extrusion step 12 (which can be done in a single machine or extruder that melts the material first, and then extrudes at least one strand). It should be noted, however, that drying could also be done in the extrusion step in accordance with German Patent Application No. 198 54 689.0 filed November 26, 1998 (Inventors: Messrs. Goedicke and Innerebner) the contents of which being incorporated here by reference.

20

It is convenient to provide a filtering step 13 either in combination with the extruding step (at the end of it, before extruding the strand) or as a separate step so as to remove impurities.

25 The extruded strand(s) are then cut to pieces or pellets in step 14. Since the material was in molten (= amorphous) condition, it is necessary to recrystallize it. Therefore, the pellets could again be fed through the precrystallizing step 2 and the crystallizing step 3, but it has been found, according to the invention, that it is sufficient to use the fluidized bed 3 having a plunger characteristic, thus shortening and economising the process.

30 Thus, it will be understood that step 3 may have a double function, i.e. one in the first production of the bottle raw material and then as a part of the reclaiming process 10 for which reason this step 3 is enveloped by a dot-dotted line 10'. This does not necessarily mean that recycling of the material is done in the same apparatus as initial production, but, in fact, this can happen.

It has been suggested in the prior art to mix the reclaimed material with fresh plastic material by melting them together. Although this can be done also when performing the process according to the present invention, it is preferred to blend the materials in the crystallizing step 3 where fresh material from step 2 can be added to reclaimed material from step 14. However, this is not critical, and it would also be possible to have a separate mixing step, e.g. before (re)condensing the material in step 4. Recondensing of reclaimed material is preferably done without any significant raise or change in intrinsic viscosity of the pellets as compared with that of the material before extrusion step 12 (Fig. 1). This means that extrusion will normally diminish the intrinsic viscosity value which is afterwards suitably raised to the range the material had before. The exact value, however, is not critical.

Fig. 2 illustrates steps 2 and 14, 3 and 4 according to a particularly preferred example. The crystallizer 3 of Fig. 1 is formed as a fluidized bed 3' where an inert gas, such as nitrogen from a source 15 of nitrogen is fed through a supply/ heater conduit 3a and through a perforated bottom 3b, while, concurrently, pellets either from cutting step 14 (Fig. 1) and/or from the precrystallizing step 2 are fed into the fluidized bed 3' via a rotary valve 3c. This bulk of pellets move substantially at uniform speed (plunger characteristic) through the fluidized bed 3' in the direction of arrow 16.

Gas, that has entered the fluidized bed 3' via conduit 3a, leaves the fluidized bed 3' at the top and via a conduit 17 leading to a cyclone 18 (or other separator, such as a filter) from which it is pumped off via a pump or fan 19. The suction power of fan 19 can be regulated by a flap (or any) valve 20. From this valve 20, a conduit 21 recycles the gas, which may have a temperature in a range of 180°C to 225°C, e.g. 205°C, to the supply/heater conduit 3a via a three-way valve V1 which can divert part of the gas stream into a conduit 22. It will be clear that conduits 17, 21 form a cycle for its own so that it would be possible to have a separate source of nitrogen for it, although it is preferred within the scope of this invention to have a common source 15 of inert gas both for the circuit 17, 21 and a further circuit to be described below. It should be noted that it is convenient to have a pressure stabilising valve 15' assigned to this source of inert gas 15. It is clear that any inert gas common in plastic production can be used.

Having moved in the sense of arrow 16 towards an outlet channel 3d of crystallizer 3', the pellets are discharged via a further rotary valve 23 into a heater/reactor 4'. In order to enable counter-current flow of gas from the reactor 4', it may be desirable to omit the rotary valve 23 entirely. In the reactor 4', the pellets are heated up to a higher temperature than in the crystallizer 3' in order to provoke reaction (condensation) of the material. This higher temperature is ordinarily above 180°C, as known *per se*. However, since the material supplied stems, at least in part, from the reclaiming process 10 (Fig. 1) it is preferred to conduct the process in a way that there is substantially no increase in iv (intrinsic viscosity) of the material as compared with that of the material before the extrusion step at 12. This means that temperatures will be maintained near the initiating point (around 185°C) of the reaction and/or the gas stream through the reactor 4' is adjusted in an appropriate way so as to avoid further increase of iv as compared with that of the material before the extrusion step 12 (Fig. 1).

It has been said that 4' designates a heater/reactor. In fact, the purpose of this apparatus is two-fold, by first preheating the pellets up to a temperature at least near the reaction (condensation) temperature so as to bring the materials temperature gently up to reaction. It has already been suggested to raise temperature in at least two steps (cf. U.S. Patent No. 3,756,990). Therefore, it can be useful, although using a single apparatus for both steps, to divide the heater/reactor 4' into two parts by introducing a heating gas at an upper annular conduit 24 communicating with the interior of the reactor 4' and a lower, similar annular conduit 25 in order to adapt gas velocity and/or quantity to the special purpose. It should, however, be noted that this will not be necessary in any case, and for some cases it would be sufficient to have a single supply over the annular conduit 25 only.

Reaction occurring internally in the material is an exothermic one. Therefore, one would suppose that the gas supplied has some cooling function and has, therefore, to be kept at a low temperature. However, what really happens within the reactor is that the internal reaction is slowed down as soon as a cloud of reaction gas surrounds every particle. Therefore, it can be the case that the reaction is accelerated and material becomes hot when too much "cooling" gas is introduced. This can result in partial melting at the top of the reactor 4', and to counter-act, it may be useful to slow down the flow of pellets in

downward direction. This can be done by braking inserts in the hollow space of the reactor 4', e.g. by providing roof-like traversing beams.

Likewise, whether the annular conduit 24 and 25 or only one is used, the gas will escape at the top, i.e. it streams as a counter-current to the downward directed flow of pellets. At the top, there is at least one exhaust conduit 26, possibly containing a pump or fan 27. Alternatively, separate exhaust conduits are provided and assigned each to the annular conduit 24 or 25 respectively. In exhaust conduit 26, there is again a three-way-valve V2 which selectively leads the stream of exhaust gas to line 17 and into the separator cyclone 18 or in a cycle to a recycling conduit 28. This recycling conduit 28 may either lead through a conduit 29 to conduit 24 and/or through a conduit 30 to conduit 25. In both conduits (or one of them) a heater 29' or 30' may be provided to establish and ensure the desired temperature which may reach 185 to 220°C and will normally be at about 200°. For using alternatively one of the conduits, it may be convenient to provide respective valves at the junctions 28' or 28" in order to regulate and direct the stream of gas properly. Furthermore, if the cycle of reactor 4' is closed over conduits 26, 28, 29 and/or 30, it will be suitable to provide another separator, similar to separator 18, just after the valve V2. Therefore, it will be apparent to those skilled in the art that it is more favourable to combine the two circuits of apparatus 3' and 4' by allowing gas to flow from conduit 26 to 17, to separator 18 and back over conduit 21 and valve V1 partially back into the crystallizer 3', partially into conduit 22 to supply gas to annular conduit 25 and, if necessary, to conduit 24 over an appropriate further valve V3.

The temperature profile along the length of the heater/reactor 4' is first increasing, then substantially maintained at a desired level and is, suitably, sharply lowered at the bottom end in order to obtain a temperature significantly below the reaction temperature, such as at 160°C. This is not necessary, but avoids the use of nitrogen (to prevent oxidising) in a following cooler 3". As is shown in Fig. 2, this cooler can be formed by a fluidized bed 3" just identical in structure as the crystallizer 3'. The difference may be only that it can have a blower 31 sucking air from the environment instead of nitrogen, if the temperature at the end of reactor 4' is significantly lowered as explained above. Therefore, the cooler 3" can have its own conduit 17' leading to separator 18'. If, however, the temperature is still in the range of the reaction temperature (at least 180°C), the circuit

of the cooler 3" could be combined with the above-mentioned circuits of apparatus 3' and 4'.

Fig. 3 shows an extruder 12a having a hopper 12b into which flakes of PET material are filled. The flakes were previously subjected to the above-mentioned steps 8, 9 and 11. During melting in the extruder 12a, gases of contaminating substances will develop and are suitably sucked off via at least one or more vacuum channels 32. The extruder 12a may be of any type known in the art. However, it is preferred to have an extruder with at least two screws, and it is even more preferred to have a plurality of screws arranged in an annular configuration when seen in cross-section.

When the plastic material leaves the extruder 12a, it is either cut into pellets immediately in the moment of passing a die by an appropriate granulator known in the art or, as may be seen in Fig. 3, after cooling in a liquid in a strand cooler 34, a cutter 35 being provided at the end of it. It should be understood that the present invention is not restricted to a special type of producing pellets.

The examples given below show how different the quality of the raw material can be. Nevertheless, a uniform bottle grade quality should be attained at the end of the process. Therefore, it is advantageous to monitor the quality (preferably the intrinsic viscosity) at a point 36a or 36b or even within the extruder 12a, by by-passing part of the molten material and analysing the by-passed amount. A vast number of methods and apparatuses are known for determining the viscosity of a material and can be used in the present process. For example, the viscosity may be determined by a NIR apparatus, as suggested at the end of a crystallizer in DE-A-43 26 105, in order to control the, particularly thermal, conditions in the subsequent steps (as indicated by dotted line 37 in Fig. 3). Other possibilities are given by using an ultra-sound detector or one of the known viscometers, such as a rotating viscometer or an on-line viscometer. Although DE-A-43 26 105 suggests already the use of a NIR viscometer, it has been found that for the purposes of reclaiming plastic material, it is useful to arrange such a viscometer 36a or 36b in a range before the crystallizer 3'.

After the possible measuring point 36b, the mixture of pellets and cooling liquid is conveniently transferred along an about S-shaped path schematically illustrated into a

separator 39 where the liquid is separated from the pellets (thus mechanically drying the surface of the pellets). Afterwards, the pellets are suitably fed to a sieve 40 to leave the pellets through (being fed to the crystallizer 3'), but to retain agglomerates of them which stick together.

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A further possibility of decontaminating the reclaimed material is using the gas circulation through at least one of the apparatuses 3', 3'' or 4' which follow the extruder/cutter unit. In Fig. 3 the gas circulation in a circuit 28a through the polycondensation reactor 4' is used by separating contaminating fines in a separator 38 (which is shown as a cyclone, but may be formed by a filter or a combination of both) and an adsorption bed 39 or a similar chemical purification apparatus. Thus volatiles and/or fines are sucked off,
10 either through the channels 32 or through the gas circuit 28a or both.

It is clear that a number of modifications are possible within the scope of the present
15 invention. For example, the type of cooler can be changed using a solid bed cooler or a belt cooler, for example. It will also be clear that whether or not all the steps indicated in Fig. 1 are done by a single producer or in different factories, it is only decisive to combine the continuous extrusion and cutting (granulating) step 12 with a continuous crystallizing step, preferably in an inert gas atmosphere, such as a nitrogen atmosphere, in
20 order to avoid all the drawbacks inherent in the prior art processes. Preferably a continuous solid state heating/condensing step is carried out subsequently.

Examples 1-3:

25 The benefits of the present invention will be better understood from the following examples. In the following Tables 1 and 2, the results of three tests are described where different amounts of plastic material were treated with an average throughput of about 50 kgs/hour. The first analyses according to Table 1 were made after extruding the material, while the results of Table 2 correspond to laboratory tests after crystallization and
30 Solid State Polycondensation (SSP).

Table 1

Example No.	Total quantity/ product	Raw material		After the extruder	
	kgs.	i.v. [dl/g]	acetaldehyde [ppm]	i.v. [dl/g]	acetaldehyde [ppm]
1	200	0.803	2.6	0.771	2.3
2	3000	0.800	5.1	0.786	2.4
3	950	0.822	3.3	0.756	2.9

- 5 Table 1 shows that the contents of acetaldehyde in the raw material can vary to a wide extent. In all tests, the intrinsic viscosity (i.v.) lowered after extrusion as could be expected. However, the contents of acetaldehyde was also lowered which illustrates a certain purification effect.
- 10 The, thus, extruded recycled material was then subjected to crystallization and condensation. The results are given in Table 2 below.

Table 2

Example No.	Total quantity/ product	After crystallization and SSP	
	kgs.	i.v. [dl/g]	acetaldehyde [ppm]
1	200	0.869	0.46
2	3000	0.884	0.46
3	950	0.848	0.48

In comparison with Table 1, Table 2 shows that the intrinsic viscosity had remarkably increased, even in comparison with that of the raw material, while the contents of acet-

aldehyde was lowered to a neglectible amount after the treatment according to the invention.

Example 4:

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In a further example, PET material to be recycled was treated in the manner described above using N₂ in the reactor 4' and a temperature of about 215°C. The time of treatment took 8 hours. The raw material and the reclaimed material at the end of the process were tested with respect to their contents of toluene, chlorobenzene, phenyl cyclohexane and benzophenone. All these contaminating substances were found to be almost removed at the end of the process, i.e. toluene with a purification efficiency of 99.4%, chlorobenzene with a purification efficiency of more than 99.9%, phenyl cyclohexane with 98.5% and benzophenone in an extent of 98.6%. This shows the high bottle grade quality obtained by the present invention.

WHAT IS CLAIMED IS:

- 5 1. A process for upgrading cleaned used plastic material to be reclaimed comprising the steps of
continuously extruding said plastic material to provide elongated strands thereof;
continuously cutting said elongated strands as they are supplied by said extrusion
step to convert them into pellets;
10 immediately feeding said pellets without intermediate storage;
continuously crystallizing said pellets, as they are fed in said feeding step from said
cutting step, in a stream of hot gas of a desired temperature to heat the pellets to
crystallizing temperature; and
solid state condensing said pellets under heat in a continuous flow after said crys-
15 tallization step to reach a desired intrinsic viscosity.
2. Process as claimed in claim 1, wherein said crystallizing step is carried out in a
continuous fluidized bed of predetermined characteristic of flow of said pellets, said
predetermined characteristic being preferably a plunger characteristic.
- 20 3. Process as claimed in claim 1 or 2, wherein at least one of the following character-
istics is provided:
a) said crystallizing step is a one-step process;
b) the step of solid state condensing said pellets is effected without substantial
25 change in intrinsic viscosity of said pellets in comparison with that of the material
before extrusion;
c) said condensing step is carried out at a temperature lower than that of said hot
gas;
d) said condensing step is carried out under a stream of gas, preferably being in
30 counter-direction to said continuous flow;
e) at least one of said steps of heating said pellets is carried out in an atmosphere
of an inert gas, preferably being nitrogen;
f) both steps of heating said pellets are carried out in a gas atmosphere, said gas
being fed from said condensing step to said crystallizing step;

g) said used plastic material comprises a polyester material, particularly polyethylene terephthalate.

4. Process as claimed in any of claims 1 to 3, further comprising the step of decontaminating the material to be reclaimed from migrated or adhering substances, said step preferably comprising sucking material off which is selected from volatiles and fines, wherein preferably the gas of at least one of said steps of heating said pellets is separated from any solid matter and is recycled.

5. Process as claimed in any of claims 1 to 4, further comprising the step of monitoring the quality of the material to be reclaimed to obtain a grading value and controlling said desired temperature of said gas or of said heat using said grading value to obtain a substantially uniform desired quality, said grading value preferably comprising the intrinsic viscosity.

6. Process as claimed in any of claims 1 to 5, further comprising at least one of the following steps:

a) cooling said pellets after said condensing step;

b) immediately feeding said pellets from said crystallizer to said condensing step without intermediate storage;

c) admixing pellets of fresh plastic material not previously used to said used plastic material, preferably comprising the step of solid state condensing said pellets in a continuous flow after said crystallization step; and

the step of cooling said pellets after said condensing step;
said admixing step being carried out after said cooling step.

7. A plant for upgrading cleaned used plastic material to be reclaimed comprising continuous extruding means for continuously extruding said plastic material; continuously operated cutting means for cutting said plastic material as they are supplied by said extrusion means to convert them into pellets; feeding means directly connected to said cutting means for feeding said pellets without intermediate storage; and

continuous crystallizing means for said pellets as they are fed from said cutting means, said crystallizing means including means creating a stream of hot gas to heat the pellets to crystallizing temperature, and preferably a continuous fluidized bed of predetermined characteristic of flow of said pellets, such as a plunger characteristic, said continuous fluidized bed, in particular, including first gas supply means for supplying a fluidisation gas for said pellets; and

continuous heating means postponed to said crystallization means for heating said pellets in a continuous flow for solid state condensation, said heating means preferably comprising second gas supply means for providing a stream of gas through said continuous flow of pellets, said stream of gas being, in particular, supplied by said second gas supply means in counter-direction to said continuous flow of pellets,

the plant suitably comprising feeding means directly connecting said crystallizing means and said heating means without intermediate storage.

8. Plant as claimed in claim 7, wherein said continuous crystallizing means comprise a continuous fluidized bed of predetermined characteristic of flow of said pellets, said continuous fluidized bed including first gas supply means for supplying a fluidisation gas for said pellets, wherein at least one of said first and second gas supply means is connected to a source of an inert gas, preferably nitrogen, said first and second gas supply means being preferably interconnected and comprising a common source of an inert gas and/or comprising recycling means for said inert gas, said recycling means comprise a duct of conduits and solids separating means in said duct of conduits.

9. Plant as claimed in claim 7 or 8, further comprising at least one of the following characteristics:

- a) cooling means arranged after said heating means to cool said pellets;
- b) fresh material supply means to at least one of said continuous means, said fresh material supply means being preferably connected to supply fresh plastic material to said crystallizing means.

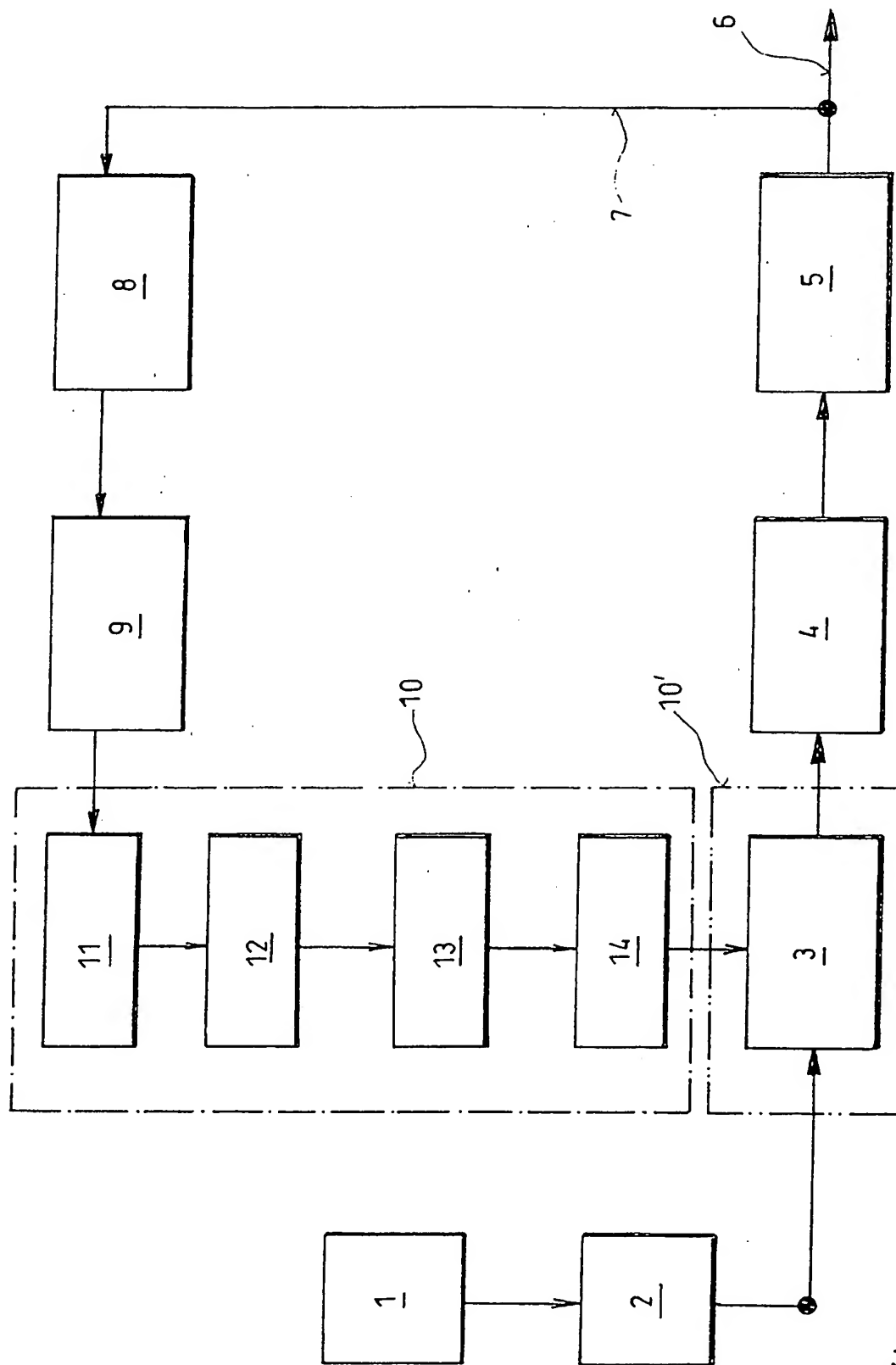


Fig.1

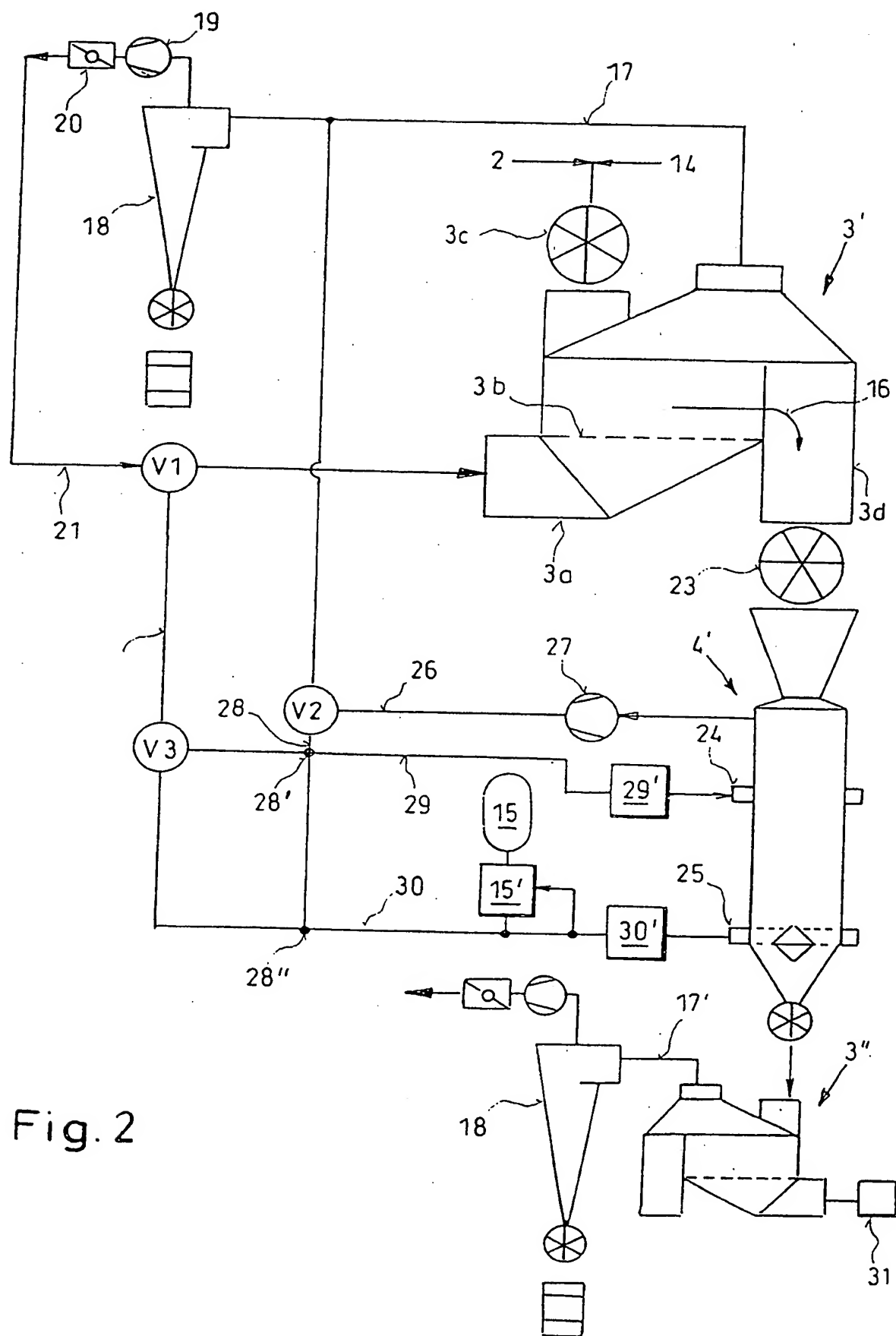


Fig. 2

3/3

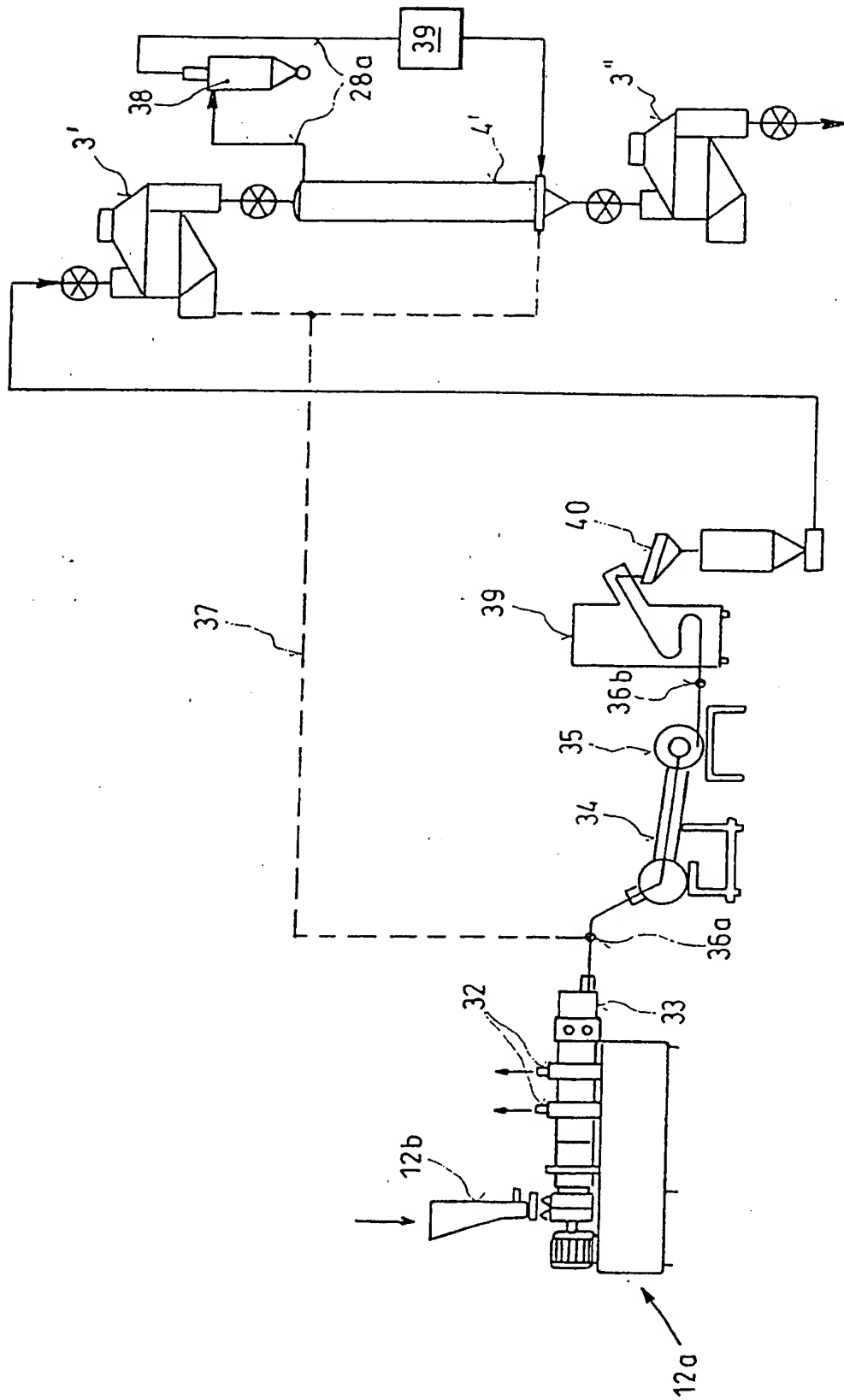


Fig. 3

INTERNATIONAL SEARCH REPORT

International Application No.

PCT/CH 99/00580

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B29B13/02 B29B17/00 B29B9/06 C08G63/80 C08J11/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B29B C08G C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 544 525 A (BALINT LASZLO J ET AL) 1 December 1970 (1970-12-01)	7-9
Y	column 1, line 15-38 column 2, line 64 column 3, line 18 column 4, line 14-28 column 5, line 4-25 column 6, line 43-59; figure 1	1-6
Y	US 5 225 130 A (DEIRINGER GUENTHER) 6 July 1993 (1993-07-06) column 5, line 13 -column 7, line 2; claim 2; figure 1	1-6
A	WO 94 01484 A (BUEHLER AG ;KAEGI WERNER (CH); RUF ARTHUR (CH); BOLLER EDWIN (CH)) 20 January 1994 (1994-01-20) figures 1,2	1-9
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28 February 2000

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 810 250 A (SINCO ENG SPA) 3 December 1997 (1997-12-03) page 1-2	1-9
A	US 5 408 035 A (DUH BEN) 18 Apr11 1995 (1995-04-18) figure 1	

INTERNATIONALER RECHERCHENBERICHT

Internationales Abkürzungszeichen

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A. KLASIFIZIERUNG DES ANMELDUNGSGEGENSTANDES

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C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie*	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
X	US 3 544 525 A (BALINT LASZLO J ET AL) 1. Dezember 1970 (1970-12-01)	7-9
Y	Spalte 1, Zeile 15-38 Spalte 2, Zeile 64 Spalte 3, Zeile 18 Spalte 4, Zeile 14-28 Spalte 5, Zeile 4-25 Spalte 6, Zeile 43-59; Abbildung 1	1-6
Y	US 5 225 130 A (DEIRINGER GUENTHER) 6. Juli 1993 (1993-07-06) Spalte 5, Zeile 13 - Spalte 7, Zeile 2; Anspruch 2; Abbildung 1	1-6
A	WO 94 01484 A (BUEHLER AG ;KAEGI WERNER (CH); RUF ARTHUR (CH); BOLLER EDWIN (CH)) 20. Januar 1994 (1994-01-20) Abbildungen 1,2	1-9

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C. (Fortsetzung) ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie*	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
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A	US 5 408 035 A (DUH BEN) 18. April 1995 (1995-04-18) Abbildung 1	

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